

(19)



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(11)

**EP 0 459 822 B1**

(12)

**EUROPEAN PATENT SPECIFICATION**

(45) Date of publication and mention  
of the grant of the patent:  
**31.01.1996 Bulletin 1996/05**

(51) Int Cl.<sup>6</sup>: **D06M 15/643, D06M 13/46**

(21) Application number: **91304942.5**

(22) Date of filing: **31.05.1991**

(54) **Dryer sheet fabric conditioner containing compatible silicones**

Konditionierungsblatt für Gewebetrockner, enthaltend kompatible Silikone

Agent de conditionnement de tissu en tant que feuille à sécher, contenant des silicones compatibles

(84) Designated Contracting States:  
**CH DE ES FR GB IT LI LU NL SE**

(30) Priority: **01.06.1990 US 532488**  
**01.06.1990 US 532430**

(43) Date of publication of application:  
**04.12.1991 Bulletin 1991/49**

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**CH DE ES FR IT LI LU NL SE**

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**US-A- 4 800 026**

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**EP 0 459 822 B1**

**Description****BACKGROUND OF THE INVENTION****FIELD OF THE INVENTION**

The instant invention relates to the application of adjuvants to fabrics in tumble-dryer automatic dryers. More particularly, it relates to an article in the form of a flexible substrate carrying a fabric conditioning composition.

**RELATED ART**

Silicones have been applied to fabrics during manufacture of fabrics or during the make up of articles of clothing. With respect to application of silicones to fabrics during a laundry process, GB-A-1,549,480; Burmeister et al., US-A-4,818,242; Konig et al., US-A-4,724,089; Konig et al., US-A-4,806,255; Dekker et al., US-A-4,661,267 and Trinh et al., US-A-4,661,269 describe aqueous dispersions or emulsions of certain silicones of limited viscosity incorporated in liquid rinse-cycle fabric softening compositions. A fabric softening composition containing emulsified silicone is also taught by Barrat et al. in US-A-4,446,033. Coffindafer et al., US-A-4,800,026 discloses fabric care compositions containing curable amine functional silicones.

The application of fabric softeners to fabrics in the tumble dryer by use of a flexible substrate carrying the fabric softeners is known in the art. The advantages of dryer added fabric conditioning include a more convenient time of addition in the laundry process and avoidance of undesirable interaction of softening agents with detergents.

Rudy et al., US-A-3,972, 131 discloses dryer sheets including a silicone oil as an ironing aid. Kasprzak et al., US-A-4,767,548 discloses the use of certain silicones in dryer sheet formulations. Coffindafer et al., US-A-4,800,026 discloses curable amine functional silicones in fabric care compositions.

In the manufacture of the dryer added fabric conditioning sheets described in the references mentioned above, when silicones are mixed with fabric softeners, the resulting mixtures are non-homogeneous and phase separation occurs readily. The homogeneity of such mixtures is ensured only by continuous vigorous agitation. An additional problem associated with the use of a nonhomogeneous mixture is the separation of actives at the point of application of the active mixture on the substrate resulting in unevenly impregnated sheets.

The compositions disclosed in the art contain individual particles of a silicone and individual particles of a fabric softening agent.

In the present invention the dispersed particle is a composite particle containing a mutually compatible mixture of a silicone and a fabric softening component. Compatible organosilicones described herein preferably form mutually soluble mixtures with certain types of commonly used fabric softening agents. Critically, the organosilicones in the dispersed composite particles do not separate from fabric softening agents during processing, on standing, during coating or drying of the dryer sheet. An additional advantage afforded by the present invention is a simplified manufacture of fabric softener compositions since the silicones need not be dispersed separately and can be introduced into the composition simultaneously with a fabric softener.

Another advantage of using compatible silicones is that compatible silicones enhance the spreading of the fabric softening agents on the fabric surface as compared to the spreading of the fabric softening agents alone or in combination with incompatible silicones. As a result of the use of compatible silicones as described herein, more complete surface coverage by a fabric softening agent is achieved with a further advantage of smaller dosage requirements. Additionally, even and uniform distribution of the actives on the dryer sheet can be attained, alleviating the problem of unevenly impregnated sheets.

Accordingly, it is a object of the invention to provide an article which provides for release of a fabric conditioning composition within an automatic laundry dryer, the composition containing a compatible mixture of a fabric softening component and a selected organosilicone.

These and other objects and advantages will appear as the description proceeds.

**SUMMARY OF THE INVENTION**

The present invention is based, in part, on the discovery that specific silicones, defined herein as compatible, are capable of forming compatible mixtures with certain conventional fabric softening agents.

It is important to differentiate between compatible and incompatible silicones and between mutually soluble and insoluble mixtures of silicones and fabric softeners. Compability as taught herein is critical and is ascertained by the appearance of the mixture of a silicone and a fabric softener. When a silicone and a fabric softener are heated and mixed together, the resulting liquid mixtures are either transparent or opaque. In the transparent mixtures, silicone and fabric softener are mutually soluble and are, accordingly suitable for use in the present invention. In the opaque mixtures,

silicone and fabric softener are mutually insoluble and the mixtures may or may not form mutually stable dispersions. A mutually stable dispersion is also compatible and is formed if a mixture of a silicone and a fabric softener does not separate into more than one phase on storage at elevated temperatures and if the mixture does not form a uniform solid or liquid on cooling. Thus, the class of compatible mixtures as defined herein includes mutually soluble mixtures as well as mutually stable dispersions. Compatibility of the mixture is critical and is determined by the silicone softener compatibility test (SSCT) described below.

In its broadest aspect, some objects of the invention are accomplished by a liquid fabric conditioning composition which includes about 1% to about 60% of composite particles containing a mutually soluble mixture of a fabric softening component and an organosilicone. Of course, these particles can also be added to a liquid containing other fabric treating ingredients, including for example, softeners.

Primarily, the invention provides an article comprising a flexible substrate carrying an effective amount of a fabric conditioning composition affixed thereto in a manner which provides for release of the conditioning composition within an automatic tumble dryer at dryer operating temperatures.

The fabric softening component employed herein for liquid compositions may be any commonly used fabric softening agent complying with the above conditions provided that it must include at least a portion of cationic quaternary ammonium salts either used singly or, optionally, in admixture with other softening agents such as nonionic softeners selected from the group of tertiary amines having at least one C<sub>8-30</sub> alkyl chain, esters of polyhydric alcohols, fatty alcohols, ethoxylated fatty alcohols, alkyl phenols, ethoxylated alkylphenols, ethoxylated fatty amines, ethoxylated monoglycerides, ethoxylated diglycerides, mineral oils, polyols, carboxylic acids having at least 8 carbon atoms, and mixtures thereof.

The fabric conditioning composition employed in articles of the present invention contains (A) certain fabric softening agents used singly or in admixture with each other and (B) an organosilicone having specific structural requirements and a specific %CH<sub>2</sub> content.

Thus, according to the primary aspect of the present invention there is provided an article for conditioning fabrics, which provides for release of a fabric conditioning composition within an automatic laundry dryer at dryer operating temperatures, the article comprising a flexible substrate carrying thereon discrete composite particles consisting of a compatible mixture of:

(a) at least 1% of a fabric softening component comprising a cationic quaternary ammonium salt; and

(b) an organosilicone of the type defined below.

#### DETAILED DESCRIPTION OF THE INVENTION

The fabric conditioning composition of the present invention includes a cationic quaternary ammonium salt. The counterion is methyl sulfate or any halide.

Examples of cationic quaternary ammonium salts include, but are not limited to:

1. Acyclic quaternary ammonium salts having at least two C<sub>8</sub> to C<sub>30</sub>, preferably C<sub>12</sub> to C<sub>22</sub> alkyl chains, such as: ditallowdimethyl ammonium chloride, di(hydrogenated tallow)dimethyl ammonium chloride, distearyldimethyl ammonium chloride, dicocodimethyl ammonium chloride and the like;

2. Cyclic quaternary ammonium salts of the imidazolinium type such as di(hydrogenated tallow)dimethyl imidazolinium methyl sulfate, 1-ethylene-bis(2-tallow-1-methyl) imidazolinium methyl sulfate and the like;

3. Diamido quaternary ammonium salts such as: methyl-bis(hydrogenated tallow amidoethyl)-2-hydroethyl ammonium methyl sulfate, methyl-bis(tallowamidoethyl)-2-hydroxypropyl ammonium methyl sulfate and the like;

4. Biodegradable quaternary ammonium salts such as N,N-di(tallowoyl-oxy-ethyl)-N,N-dimethyl ammonium chloride, and N,N-di(tallowoyl-oxy-propyl)-N,N-dimethyl ammonium chloride and the like. When fabric conditioning compositions employ biodegradable quaternary ammonium salts, the pH of the composition is preferably adjusted to between about 2 and about 5. Biodegradable quaternary ammonium salts are described, for example, in US-A-4,767,547 and US-A-4,789,491.

5. Mixtures of water-insoluble cationic fabric softeners and polyalkoxylated ammonium salts as described in US-A-4,422,949. Such mixtures are particularly suitable for incorporation in concentrated form of the liquid compositions herein.

The fabric softening component may include other fabric softeners in addition to the cationic quaternary ammonium salts. Additional fabric softeners suitable for use herein can be selected from the following classes of compounds:

i. Tertiary fatty amines having at least one and preferably two  $C_8$  to  $C_{30}$ , preferably  $C_{12}$  to  $C_{22}$  alkyl chains. Examples include hardened tallow amine and cyclic amines such as 1-(hydrogenated tallow)amidoethyl-2-(hydrogenated tallow) imidazoline. Cyclic amines which may be employed for the compositions herein are described in US-A-4,806,255.

ii. Carboxylic acids having 8 to 30 carbon atoms and one carboxylic group per molecule. The alkyl portion has 8 to 30, preferably 12 to 22 carbon atoms. The alkyl portion may be linear or branched, saturated or unsaturated, with linear saturated alkyl preferred. Stearic and myristic acids are preferred fatty acids for use in the composition herein. Examples of these carboxylic acids are commercial grades of stearic acid and the like which may contain small amounts of other acids.

iii. Esters of polyhydric alcohols such as sorbitan esters or glycerol stearate. Sorbitan esters are the condensation products of sorbitol or iso-sorbitol with fatty acids such as stearic acid. Preferred sorbitanesters are monoalkyl. A common example of sorbitan ester is SPAN 60 (ICI) which is a mixture of sorbitan and isosorbide stearates.

iv. Fatty alcohols, ethoxylated fatty alcohols, alkylphenols, ethoxylated alkyl phenols, ethoxylated fatty amines, ethoxylated monoglycerides and ethoxylated diglycerides.

v. Mineral oils, and polyols such as polyethylene glycol.

vi. Condensation products of higher fatty acids with polyamines, selected from the group consisting of hydroxyalkyl alkylene diamines, dialkylene triamines and mixtures thereof, as described in US-A-4,661,269.

Preferred fabric softeners for use herein are acyclic quaternary ammonium salts, ditallowdimethyl ammonium chloride being most preferred for fabric conditioning compositions of this invention. Especially preferred are mixtures of ditallowdimethyl ammonium chloride with fatty acids, particularly stearic acid or myristic acid.

About 1% to about 40% of the fabric softening component is typically used in the compositions of the invention. There must be included at least a sufficient amount of quaternary ammonium salt to achieve anti-static effect, for example, about 1% to 3% in the dilute product and about 2% to about 5% in the concentrated product. On the other hand, the entire fabric softening component may be quaternary ammonium salt. The diluted version of the product contains about 1% to about 12%, preferably about 3% to about 10% and most preferably about 4% to about 7% of the fabric softening component. The concentrated version of the product contains about 13% to about 40%, preferably about 13% to 30% and most preferably about 13% to about 20% of the fabric softening component.

When the fabric softening composition is carried on a flexible substrate according to the article of the present invention the fabric softening agents (A) include conventionally used cationic and nonionic fabric softening agents such as those listed in (1) to (5) and (i) to (vi) above.

The amount of the fabric softening composition on the sheet is subject to normal coating parameters such as, for example, viscosity and melting point of the fabric softening components and is typically about 0.5 grams to about 5 grams, preferably about 1 gram to about 3.5 grams. The fabric softening composition employed in the present invention contains at least 1%, up to about 95% of the fabric softening component. Preferably from about 10% to about 80% and most preferably from about 30% to about 70% of the fabric softening component is employed herein to obtain optimum softening at minimum cost.

The quaternary ammonium salt is typically used in the amount of about 10% to about 80%, preferably about 30% to about 70%.

#### Silicone

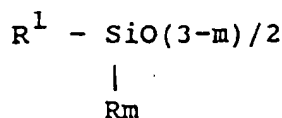
The second essential ingredient of the fabric softening composition employed in the present invention is a compatible organosilicone.

The organosilicones employed in the present invention (also termed herein as compatible silicones) are capable of forming compatible mixtures with the fabric softeners listed above.

The organosilicones employed herein have a  $\%CH_2$  content of about 25% to about 90%. The  $\%CH_2$  content is defined as

$$\%CH_2 = \frac{\text{number of methylene (CH}_2\text{) groups}}{\text{number of methylene groups and methyl groups}} \times 100\%$$

The organosilicones included in the fabric conditioning compositions of the invention contain at least one unit of Formula A:

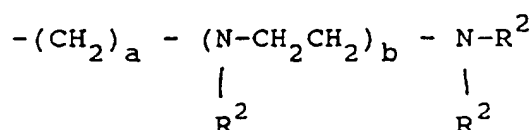


wherein m is a number from 0 to 2 and R is a mono valent hydrocarbon radical.

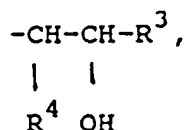
The value of (3-m)/2 in Formula A means the ratio of oxygen atoms to silicon atoms, i.e. SiO<sub>1/2</sub> means one oxygen is shared between two silicon atoms.

R<sup>1</sup> in Formula A is selected from the group consisting of:

i. a unit of Formula A1

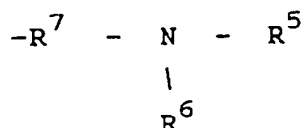


wherein a is a number of at least 1, preferably 3; b is a number from 0 to 10, preferably 1; R<sup>2</sup> is

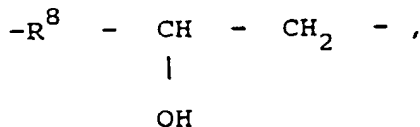


R<sup>3</sup> is a hydrocarbon radical having from 4 to 40 carbon atoms preferably from 8 to 18 carbon atoms and may be saturated, unsaturated, cyclic, acyclic, alkyl or aromatic; and R<sup>4</sup> is hydrogen or a hydrocarbon radical having from 1 to 40 carbon atoms, preferably hydrogen; and

ii. a unit of Formula A2



wherein R<sup>5</sup> and R<sup>6</sup> are independently selected from hydrogen or a hydrocarbon radical having from 1 to 45 carbon atoms which may be saturated, unsaturated, cyclic, acyclic, alkyl or aromatic and at least one of R<sup>5</sup> and R<sup>6</sup> is a hydrocarbon radical having from 6 to 45 carbon atoms, R<sup>7</sup> is



wherein R<sup>8</sup> is a divalent organic radical having from 1 to 12 carbon atoms which may be saturated, unsaturated, cyclic, acyclic, alkyl or aromatic, and preferably is -CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>-O-CH<sub>2</sub>.

Thus, organosilicones employed in the present invention include alkylaminosilicones which satisfy the structural parameters described above and which have % methylene (%CH<sub>2</sub>) content of about 25% to about 90%. Compatibility of the organosilicones herein with fabric softening agents depends, in part, on the %CH<sub>2</sub> content of the organosilicones. The preferred range of the %CH<sub>2</sub> content for the silicones herein is from about 40% to about 90%, more preferably from about 50% to about 85%, and most preferably from about 50% to about 75% to increase the degree of compatibility of the mixtures containing relatively large amounts of silicone.

The organosilicones included in the compositions herein may be linear, branched, or partially crosslinked, preferably



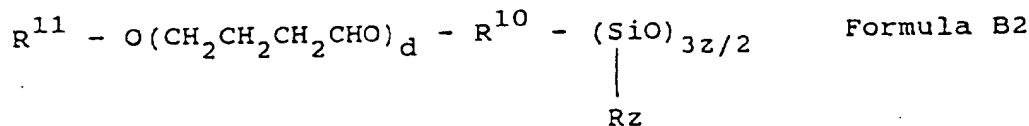
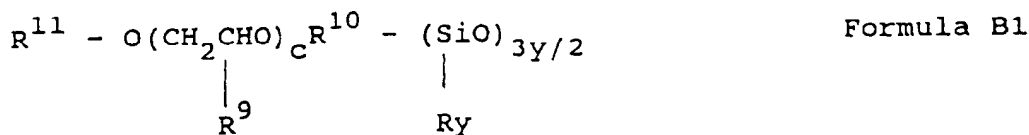
Formula A1, or 2) by treating epoxysilicones with primary or secondary amines such as dicocoamine to form alkylaminosilicones having the unit of Formula A2.

The modified alkylaminosilicones of the invention having the unit of Formula A1 may be prepared by mixing epoxide compounds with aminosilicones in a pressure reactor and heating for about 24 hours, after which the unreacted epoxide compound is vacuum stripped off. The amount of epoxide to be used is calculated based upon the number of amine functional groups on the alkylaminosilicone. Preferably, two epoxides are reacted for every primary amine and one epoxide for every secondary amine, in order to convert them to tertiary amines. A stoichiometric amount or up to 25% excess of epoxide can be used. The reaction is preferably conducted between 25°C and 150°C, especially between 50°C and 100°C. The pressure is preferably maintained from  $3.447 \times 10^5 \text{ Nm}^{-2}$  (50 psi) to  $2.068 \times 10^6 \text{ Nm}^{-2}$  (300 psi), particularly from  $3.447 \times 10^5 \text{ Nm}^{-2}$  (50 psi) to  $1.034 \times 10^6 \text{ Nm}^{-2}$  (150 psi). Typical aminosilicone starting compounds would include Dow Corning Q2-8075. The art of making alkylaminosilicones having the unit of Formula A1 is disclosed in Examples 1 and 2 herein and in the US-A-4 994 593 Lin et al. entitled 'Hydroxylhydrocarbyl Modified Aminoalkyl Silicones', filed December 6, 1989.

The modified alkylaminosilicones having the unit of Formula A2 may be prepared by mixing epoxysilicones, secondary amines, and a solvent such as isopropanol or toluene, and heating the mixture at reflux for about 24 hours, after which the solvent is removed by distillation or vacuum stripping. The amount of amine to be used is calculated based upon the number of epoxy functional groups on the epoxysilicone. Preferably, one secondary amine is reacted for every epoxy functional group in order to convert the amine to tertiary amine. A stoichiometric amount or up to 25% excess of amine can be used. The reaction is preferably conducted between 50°C and 150°C, especially between 75°C and 110°C. The reaction is preferably conducted at atmospheric pressure, but may be conducted in a pressure reactor with the pressure being maintained from  $3.447 \times 10^5 \text{ Nm}^{-2}$  (50 psi) to  $2.068 \times 10^6 \text{ Nm}^{-2}$  (300 psi).

The modified alkylaminosilicones employed in this invention contain amine groups which may be quaternised with, for example, alkyl halide or methyl sulfate, or may be protonated with a Lewis acid such as hydrochloric acid, acetic acid, citric acid, formic acid and the like.

Alkylaminosilicones employed herein may, in addition to the units of Formula A, contain secondary units selected from the group consisting of a unit of Formula B1 and a unit of Formula B2:



wherein  $\text{R}^{11}$  is a hydrocarbon radical having from 1 to 40 carbon atoms, preferably is  $\text{CH}_3$ ;  $\text{R}^9$  is a hydrocarbon radical having from 1 to 3 carbon atoms;  $\text{R}^{10}$  is oxygen or alkylene having from 1 to 8 carbon atoms, preferably propylene; c and d are numbers from 0 to 50, preferably 2 to 15; and y and z are numbers from 0 to 2.

Organosilicones preferred for use herein have a  $\% \text{CH}_2$  content of about 40% to about 90% and are alkylaminosilicones having the unit of Formula A1.

The amount of organosilicone employed herein generally ranges from about 0.1% to about 20%, and is preferably at least about 0.5% to about 2% to maximise the spreading of the fabric softeners on fabric surface, but could be higher in concentrated liquids. Preferably when the organosilicone is carried on a substrate, the amount employed is from 0.1% to 20%, more preferably at least 3% to 20%. The amount of the organosilicone is governed by the ratio at which the mutually soluble mixture of the fabric softening component and the organosilicone is formed.

The weight ratio of the organosilicone to the fabric softening component in the fabric conditioning compositions employed herein is from about 100:2 to about 1:100, preferably from about 2:100 to about 20:100, but must be such that a compatible mixture can be formed.

#### Silicone/Softener Compatibility Test (SSCT)

As described above, mixtures defined as compatible herein include mutually soluble as well as mutually stable dispersible mixtures. Compatibility of the fabric conditioning mixtures herein depends on the structure and the  $\% \text{CH}_2$  content of the organosilicone and the particular fabric softeners employed in the mixture. SSCT provides a basis for selecting appropriate combinations of the fabric softening component and the organosilicone.

The test may be used to determine the compatibility at a particular weight ratio of interest or to determine a minimum concentration of the silicone at which a compatible mixture of the silicone and the fabric softening component is formed.

SSCT is conducted as follows:

a 10 gram sample of the fabric softener or a combination of fabric softeners is placed into a clear glass flask equipped with a stirring mechanism, such as a magnetic stirrer. If either the fabric softener or the silicone is a solid at room temperature, it is melted before the test is begun with the test taking place above the melting point of the fabric softener or the silicone. The silicone of interest is slowly introduced with, conveniently, a Pasteur pipet into the flask, with stirring. It is estimated that the weight of one drop represents about 1% silicone concentration, so the silicone is mixed with the fabric softener 1% at a time. Thus, the lowest concentration of the silicone in the mixture is about 1%.

If the resulting mixture of the fabric softening agent and the silicone stays clear over the entire investigated range of the silicone, this indicates that the components of the mixture are mutually soluble over the investigated concentration range and, accordingly, are compatible. Clear mixtures are defined herein as mixtures having about 90% transmittance when measured with a visible light probe (one centimeter pathlength) against distilled water background using Brinkman PC800 colormeter.

The mixture may also become cloudy indicating that the silicone and the fabric softener are not mutually soluble at that weight % of the silicone. In this case, if the mixture became cloudy, the weight percent of the silicone added to produce cloudiness is calculated. This number, termed compatibility  $\alpha$ , then represents the weight percent of the silicone to produce a cloudy mixture. Cloudy samples are placed in an oven at 100°C for at least two hours, then cooled to room temperature and inspected. Samples which have completely separated into distinct layers are incompatible and are not useful for the invention. Samples which maintain a stable, dispersed character are compatible and, hence, useful in the invention.

It is sufficient, for practical applications, to investigate the silicone concentration range of up to about 30%. However, the entire range up to 100% of the silicone concentration may be investigated if desired. When the entire range of the silicone concentration is to be investigated, the silicone is added until the mixture contains about 60% by weight of the silicone. Silicone addition is then stopped, and the experiment is repeated by adding the fabric softener to a 10 gram sample of the silicone. In those samples that became cloudy, the weight percent of the softener added to produce cloudiness is calculated and subtracted from 100, the resulting number is termed herein compatibility  $\beta$ .

$\alpha$  compatibility reflects compatibility of the mixtures containing a fabric softener as a major component, whereas  $\beta$  compatibility reflects compatibility of the mixtures containing a silicone as a major component. Minimal difference between  $\beta$  and  $\alpha$  ( $\beta - \alpha$ ) reflects degree of compatibility of the mixture: more compatible mixtures have a lower number for  $\beta - \alpha$ .

Preferably, the silicone and the fabric softening component are compatible at a silicone concentration of at least about 2%.

Mutually soluble and clear mixtures of the silicone and the fabric softening component indicate the highest degree of compatibility and are preferred.

Various additives may be used in combination with the compatible mixture of the fabric softening component and the compatible silicone. The additives are used in the amounts that do not substantially affect the compatibility of the mixture and include small amounts of incompatible silicones, such as predominantly linear polydialkylsiloxanes, e.g. polydimethylsiloxanes; soil release polymers such as block copolymers of polyethylene oxide and terephthalate; fatty amines selected from the group consisting of primary fatty amines, secondary fatty amines, tertiary fatty amines and mixtures thereof; amphoteric surfactants; smectite type inorganic clays; anionic soaps, switterionic quaternary ammonium compounds; and nonionic surfactants.

Other optional ingredients include emulsifiers, electrolytes, optical brighteners or fluorescent agents, buffers, perfumes, colourants, germicides and bactericides.

The fabric conditioning compositions of the invention can be used in the rinse cycle of a conventional home laundry operation. Generally, rinse water has a temperature of from about 5°C to about 70°C. The concentration of the total active ingredients is generally from about 2 ppm to about 1000 ppm, preferably from about 10 ppm to about 500 ppm, by weight of the aqueous rinsing bath. When multiple rinses are used, the fabric conditioning composition is preferably added to the final rinse.

However, according to the primary aspect of the invention, an article is disclosed for conditioning fabrics in a tumble drier. The article of the invention comprises a flexible substrate which carries a fabric conditioning amount of a conditioning composition and is capable of releasing the conditioning composition at dryer operating temperatures. The conditioning composition in turn has a preferred melting (or softening) point of about 25°C to about 150°C.

The fabric conditioning composition employed in the invention is coated onto a dispensing means which effectively releases the fabric conditioning composition in a tumble dryer. Such dispensing means can be designed for single usage or for multiple uses. One such article comprises a sponge material releasably enclosing enough of the conditioning composition to effectively impart fabric softness during several drying cycles. This multi-use article can be made by filling a porous sponge with the composition. In use, the composition melts and leaches out through the pores of the sponge to soften and condition fabrics. Such a filled sponge can be used to treat several loads of fabrics in conventional



dryers, and has the advantage that it can remain in the dryer after use and is not likely to be misplaced or lost.

Another article comprises a cloth or paper bag releasably enclosing the composition and sealed with a hardened plug of the mixture. The action and heat of the dryer opens the bag and releases the composition to perform its softening.

A highly preferred article comprises the compositions containing a softener and a compatible organosilicone releasably affixed to a flexible substrate such as a sheet of paper or woven or nonwoven cloth substrate. When such an article is placed in an automatic laundry dryer, the heat, moisture, distribution forces and tumbling action of the dryer removes the composition from the substrate and deposits it on the fabrics.

The sheet conformation has several advantages. For example, effective amounts of the compositions for use in conventional dryers can be easily absorbed onto and into the sheet substrate by a simple dipping or padding process. Thus, the end user need not measure the amount of the composition necessary to obtain fabric softness and other benefits. Additionally, the flat configuration of the sheet provides a large surface area which results in efficient release and distribution of the materials onto fabrics by the tumbling action of the dryer.

The substrates used in the articles can have a dense, or more preferably, open or porous structure. Examples of suitable materials which can be used as substrates herein include paper, woven cloth, and non-woven cloth. The term 'cloth' herein means a woven or non-woven substrate for the articles of manufacture, as distinguished from the term 'fabric' which encompasses the clothing fabrics being dried in an automatic dryer.

It is known that most substances are able to absorb a liquid substance to some degree; however, the term 'absorbent', as used herein, is intended to mean a substrate with an absorbent capacity (i.e., a parameter representing a substrate's ability to take up and retain a liquid) from 4 to 12, preferably 5 to 7 times its weight of water.

If the substrate is a foamed plastics material, the absorbent capacity is preferably in the range of 15 to 22, but some special foams can have an absorbent capacity in the range from 4 to 12.

Determination of absorbent capacity values is made by using the capacity testing procedures described in U.S. Federal Specification (UU-T-595b), modified as follows:

1. tap water is used instead of distilled water;
2. the specimen is immersed for 30 seconds instead of 3 minutes;
3. draining time is 15 seconds instead of 1 minute; and
4. the specimen is immediately weighed on a torsion balance having a pan with turned-up edges.

Absorbent capacity values are then calculated in accordance with the formula given in said Specification. Based on this test, one-ply, dense bleached paper (e.g., Kraft or bond having a basis weight of about 14.52 kg (32 pounds) per  $2,787 \times 10^2 \text{ m}^2$  (3,000 square feet)) has an absorbent capacity of 3.5 to 4; commercially available household one-ply towel paper has a value of 5 to 6; and commercially available two-ply household towelling paper has a value of 7 to about 9.5.

Suitable materials which can be used as a substrate in the invention herein include, among others, sponges, paper, and woven and non-woven cloth, all having the necessary absorbency requirements defined above.

The preferred non-woven cloth substrates can generally be defined as adhesively bonded fibrous or filamentous products having a web or carded fibre structure (where the fibre strength is suitable to allow carding), or comprising fibrous mats in which the fibres or filaments are distributed haphazardly or in random array (i.e. an array of fibres in a carded web wherein partial orientation of the fibres is frequently present, as well as a completely haphazard distribution orientation), or substantially aligned. The fibres or filaments can be natural (e.g. wool, silk, jute, hemp, cotton, linen, sisal, or ramie) or synthetic (e.g. rayon, cellulose ester, polyvinyl derivatives, polyolefins, polyamides, or polyesters).

The preferred absorbent properties are particularly easy to obtain with non-woven cloths and are provided merely by building up the thickness of the cloth, i.e., by superimposing a plurality of carded webs or mats to a thickness adequate to obtain the necessary absorbent properties, or by allowing a sufficient thickness of the fibres to deposit on the screen. Any diameter or denier of the fibre (generally up to about 10 denier) can be used, inasmuch as it is the free space between each fibre that makes the thickness of the cloth directly related to the absorbent capacity of the cloth, and which, further, makes the non-woven cloth especially suitable for impregnation with a composition by means of inter-sectional or capillary action. Thus, any thickness necessary to obtain the required absorbent capacity can be used.

When the substrate for the composition is a non-woven cloth made from fibres deposited haphazardly or in random array on the screen, the articles exhibit excellent strength in all directions and are not prone to tear or separate when used in the automatic clothes dryer.

Preferably, the non-woven cloth is water-laid or air-laid and is made from cellulosic fibres, particularly from regenerated cellulose or rayon. Such non-woven cloth can be lubricated with any standard textile lubricant. Preferably, the fibres are from 5mm to 50mm in length and are from 1.5 to 5 denier. Preferably, the fibres are at least partially orientated

haphazardly, and are adhesively bonded together with a hydrophobic or substantially hydrophobic binder-resin. Preferably, the cloth comprises about 70% fibre and 30% binder resin polymer by weight and has a basis weight of from about 18 to 45g per square metre.

In applying the fabric conditioning composition to the absorbent substrate, the amount impregnated into and/or coated onto the absorbent substrate is conveniently in the weight ratio range of from about 10:1 to 0.5:1 based on the ratio of total conditioning composition to dry, untreated substrate (fibre plus binder). Preferably, the amount of the conditioning composition ranges from about 5:1 to about 1:1, most preferably from about 3:1 to 1:1, by weight of the dry, untreated substrate.

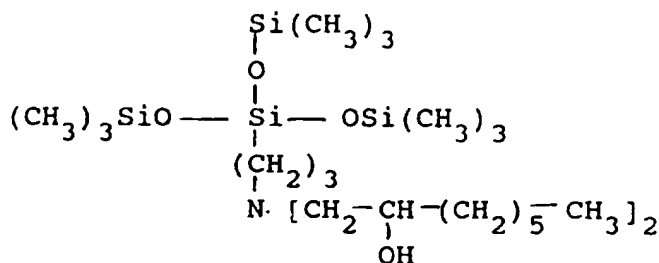
According to one preferred embodiment of the invention, the dryer sheet substrate is coated by being passed over a rotogravure applicator roll. In its passage over this roll, the sheet is coated with a thin, uniform layer of molten fabric softening composition contained in a rectangular pan at a level of about 17.94 g/m<sup>2</sup> (15g/square yard). Passage of the substrate over a cooling roll then solidifies the molten softening composition to solid. This type of applicator is used to obtain a uniform homogeneous coating across the sheet.

Following application of the liquefied composition, the articles are held at room temperature until the composition substantially solidifies. The resulting dry articles, prepared at the composition substrate ratios set forth above, remain flexible; the sheet articles are suitable for packaging in rolls. The sheet articles can optionally be slitted or punched to provide a non-blocking aspect at any convenient time if desired during the manufacturing process.

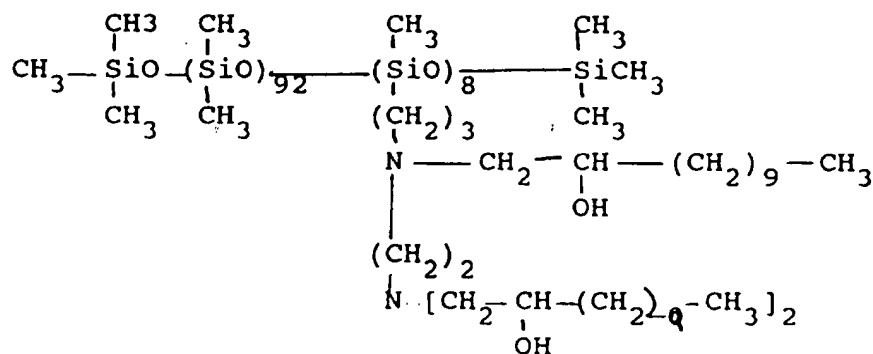
The following Examples will more fully illustrate the embodiments of this invention. All parts, percentages and proportions referred to herein and in the appended claims are by weight of the composition unless otherwise indicated.

Examples 1-6 include organosilicones within the scope of the present invention having formulas A, B, C and D:

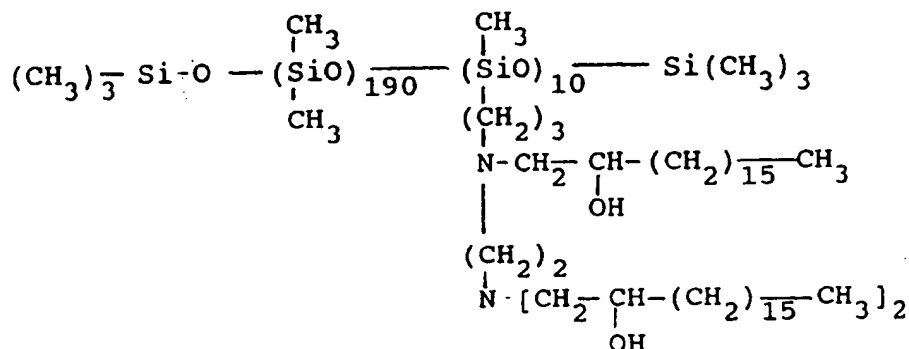
### Formula A



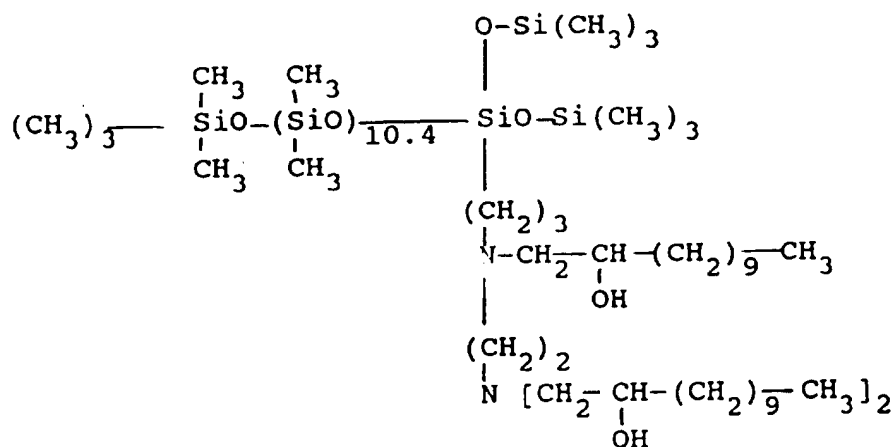
### Formula B



**Formula C**



### Formula D



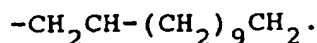
### EXAMPLE 1

The silicone of Formula C is a reaction product of the starting aminosilicone (where the nitrogen-containing branch chain is  $-(CH_2)_3-NH-(CH_2)_2NH_2$ ) and 1,2 epoxyoctadecane. The compound was prepared by placing the starting aminosilicone (61.16g), 1,2 epoxyoctadecane (38.84g) and 2-propanol (60.0g) in a reaction vessel and heating to 80°C for 24 hours. The reaction vessel consisted of a three neck round bottom flask containing a stirrer, a reflux condenser and a thermometer. The 2-propanol was then stripped off with a  $N_2$  sparge at 100°C as described in the Lin et al. application mentioned above.

Formula C silicone has %CH<sub>2</sub> equal 56.62.

### EXAMPLE 2

A 'T' structure modified alkylaminosilicone of Formula D, having %CH<sub>2</sub> equal 52.50 was prepared. In the starting aminoalkylsilicone, the nitrogen-containing branch chain is -(CH<sub>2</sub>)<sub>3</sub>-NH-(CH<sub>2</sub>)<sub>2</sub>NH<sub>2</sub>. In the modified aminoalkylsilicone hydrogens on nitrogens were replaced with



OH

In the process, 34.7g of the starting aminoalkylsilicone, 34.4g 1,2-epoxydodecane and 17.4g 2-propanol were charged to the reaction vessel following the procedures of Example 1.

EXAMPLE 3

Effect of the %CH<sub>2</sub> content of various silicones as indicated in Table I on the compatibility with Adogen 442 (dihydrogenated tallow dimethyl ammonium chloride from Sherex Corp.) was investigated by mixing the silicones with Adogen 442, following the SSCT procedure.

The results that were generated are summarised in Table I. Samples 6 and 7 were synthesised in Examples 1 and 2 respectively.

Table I

| #  | Silicone              | %CH <sub>2</sub> | Compatible |
|----|-----------------------|------------------|------------|
| 1. | DC 200 <sup>1</sup>   | 0                | no         |
| 2. | DC SSF <sup>2</sup>   | 0                | no         |
| 3. | Formula A             | 56.69            | yes        |
| 4. | Formula B             | 57.61            | yes        |
| 5. | Formula B, protonated | 57.61            | yes        |
| 6. | Formula C             | 56.62            | yes        |
| 7. | Formula D             | 52.50            | yes        |

<sup>1</sup>Linear polydimethylsiloxane, supplied by Dow Corning, viscosity =  $1 \times 10^{-3} \text{ m}^2 \text{ s}^{-1}$  (1000cst)

<sup>2</sup>Aminosilicone supplied by Dow Corning, amine neutral equivalent = 2000, viscosity =  $1.3 \times 10^{-4} \text{ m}^2 \text{ s}^{-1}$  (130cst).

Silicones of samples 3 - 7 were mutually soluble with Adogen 442 at silicone concentration of 5% by weight of the mixture. However, silicone 1 and 2, which are not within the scope of the present invention, were not compatible with Adogen 442 at 5% or even at 25% of silicone.

Examples 4-6Contact Angle Measurements

Contact angle values reflect the spreading behaviour of a liquid on a solid surface. Discussion of the relationship between contact angle values and spreading is provided, for example, in Chapter 6 of "Introduction to Colloid and Surface Chemistry", Duncan J. Shaw, Butterworth, 1985. A contact angle of a liquid on solid surface is the angle between the tangent of the droplet and the surface. A smaller contact angle indicates better spreading on the surface. When it is desired to measure the contact angle on fabrics, there is an experimental problem of accurately measuring the true contact angle: due to the surface roughness of the fabric it is difficult to obtain an accurate baseline. Thus, the true contact angle measurements were obtained using cellulose paper.

Samples were prepared by mixing a fabric softener and a silicone above the melting point. A droplet of the melt liquid was applied to a piece of cellulose filter paper. After the droplet cooled and solidified, an initial contact angle was measured. The cellulose paper with the droplet was then placed in a 70°C oven for 30 minutes in order for the equilibrium contact angle to be achieved. The paper was then removed from the oven and a final contact angle was measured.

The contact angle was measured using a contact angle goniometer (Rame'-Hart model 100). The cellulose with the drop of active was placed on the stage and viewed with a microscope. With the light source on, the drop appeared as a silhouette against a soft, green background. The drop/cellulose interface was aligned with the horizontal crosshair, and the contact angle was determined by rotating the read-out crosshair to tangency with the drop right profile. The contact angle value was then read directly on the graduated goniometer scale. This procedure was repeated to read the contact angle on the left side. Both sides should give the same reading otherwise the sample was not levelled

correctly and the stage height should be readjusted.

#### Example 4

Effect of various silicones as indicated in Table II on the spreading of Adogen 442 was investigated. The true contact angle (initial and final) of the mixtures of silicones and Adogen 442 prepared in Example 3 was measured on cellulose paper as described above. Additionally, spreading of the mixtures on cotton fabric was evaluated qualitatively, using a score of 1 to 4: 1 = best spreading, 2 = moderate spreading, 3 = droplet starting to wet the surface, 4 = no spreading, droplet beading up. Sample 1 contained only Adogen 442 without any silicone and was used as a control.

The results that were generated are summarised in Table II.

TABLE II

| Sample No. | Silicone              | Cotton | Cellulose |       |
|------------|-----------------------|--------|-----------|-------|
|            |                       |        | Initial   | Final |
| 1          | none                  | 4      | 110       | 112   |
| 2          | DC200                 | 4      | 110       | 147   |
| 3          | DCSSF                 | 4      | 95        | 132   |
| 4          | Formula A             | 1      | 70        | 18    |
| 5          | Formula B             | 2-3    | 72        | 59    |
| 6          | Formula B, protonated | 2      | 59        | 21    |
| 7          | Formula C             | 2-3    | 86        | 57    |
| 8          | Formula D             | 1      | 47        | 60    |

Initial and final contact angles for samples 4-8 containing compatible silicones within the scope of the invention were lower than contact angles for samples 1-3.

Silicones of samples 4-8 were shown to form mutually soluble mixtures with Adogen 442 in Example 3.

Samples 1-3 contained either no silicone or silicones which are not within the scope of the invention. The results established that, in mutually soluble mixtures of compatible silicones and fabric softener as taught by the present invention, compatible silicones improve the spreading of the fabric softener on a cellulose surface. Qualitative evaluation of spreading on cotton showed the same pattern of improved spreading when compatible silicones within the scope of the invention were used.

#### Example 5

The concentration effect of various silicones as indicated in Table III on the spreading of Adogen 442 fabric softener was investigated by measuring the contact angle on a cellulose surface using the procedure described above.

TABLE III

| Sample No. | Silicone              | Final Contact 1.5% | Angle at 3.5% | % Silicone of 7.5% |
|------------|-----------------------|--------------------|---------------|--------------------|
| 1          | DC200                 | 147                | 147           | 147                |
| 2          | Formula B             | 118                | 55            | -                  |
| 3          | Formula B, protonated | 48                 | 20            | 20                 |
| 4          | Formula D             | 42                 | -             | 55                 |
| 5          | Formula D, protonated | 98                 | 78            | 5                  |

This example demonstrates that in Samples 2-5 containing organosilicones within the scope of the invention as little as about 2% by weight of the mixture is needed to reduce the contact angle to improve the spreading on the surface.

Further increase in silicone concentration in Samples 2-5 further reduced the contact angle, indicating even better spreading on the surface.

Silicone of sample 1 which is not suitable for the present invention did not reduce the contact angle of the fabric softener regardless of the amount of the silicone used.

EXAMPLE 6

Mixtures of various silicones as indicated in Table IV with nonionic fabric softeners, such as mineral oil were investigated. The spreading of the mixtures on cotton and polycotton fabrics was investigated by measuring the fabric area (centimetres<sup>2</sup>) per gram of mineral oil spread on the fabrics.

All samples contained 5% by weight of the mixture of a silicone. The mineral oil used was Semsol 350 from Witco Corp.

TABLE IV

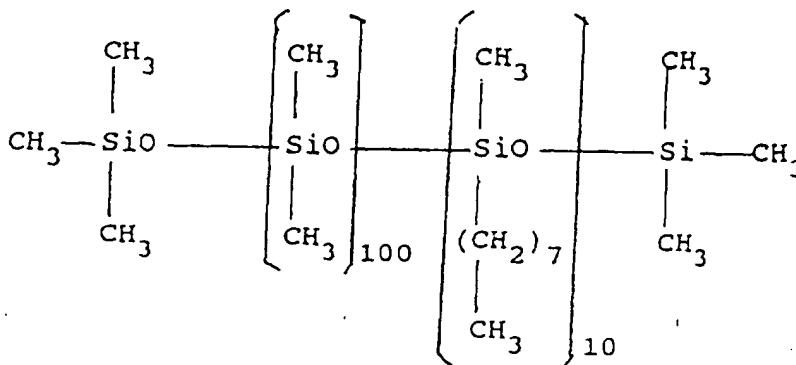
| Sample No. | Silicone  | Viscosity (m <sup>2</sup> s <sup>-1</sup> /cst) | Surface Tension (dyne/cm) | Fabric Area |            |
|------------|-----------|---|---------------------------|-------------|------------|
|            |           |   |                           | Cotton      | Polycotton |
| 1          | none      | 1.05 x 10 <sup>-4</sup> /105                    | 32.0                      | 303         | 371        |
| 2          | Formula B | 2.95 x 10 <sup>-4</sup> /295                    | 22.9                      | 227         | 224        |
| 3          | Formula D | 1.82 x 10 <sup>-4</sup> /182                    | 22.2                      | 326         | 552        |

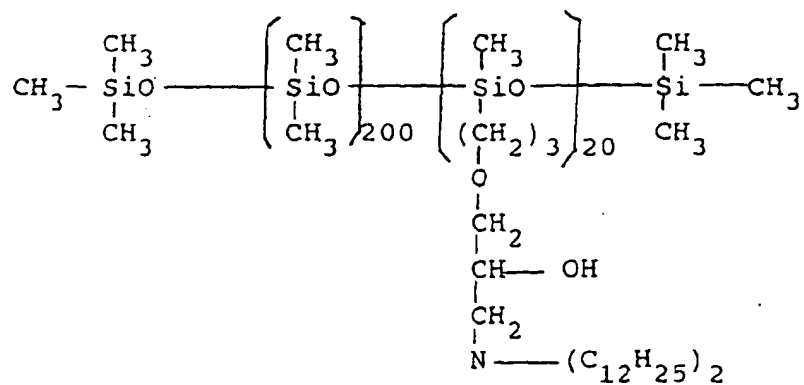
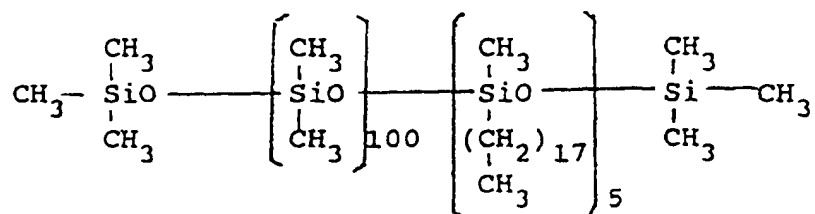
Formula B silicone was only partially soluble in mineral oil, while Formula D silicone formed a mutually soluble mixture with mineral oil, demonstrating that the mutual solubility of the silicones and fabric softeners depend on the particular fabric softener as well as the %CH<sub>2</sub> of the silicone.

Silicones B and D both reduced the surface tension of mineral oil as observed in the absence of silicones in sample 1. However, fabric area coverage was increased only in sample 3 where a mutually soluble mixture was formed.

EXAMPLES 7-8

Examples 7-8 include organosilicones both outside and within the scope of the invention having formulas E, F and G.

Formula E (comparison example)

Formula FFormula G (comparison example)EXAMPLE 7

The mutual solubility of organosilicones with mixtures of fabric softening agents was investigated in the following formulations:

| Formulation<br>No. | Fabric Softening<br>Component Mixture |
|--------------------|---------------------------------------|
|--------------------|---------------------------------------|

|     |  |
|-----|--|
| I   | 10% Varisoft 475 <sup>1</sup><br>10% Mineral Oil     |
| II  | 10% Adogen 442<br>1% Myristic Acid                   |
| III | 11.7% Varisoft 445 <sup>2</sup><br>3.5% Stearic Acid |

<sup>1</sup>Varisoft 475 = Methyl-1-tallowamidoethyl-2-tallow  
imidazolinium methyl sulfate

<sup>2</sup>Varisoft 445 = Methyl-1-hydrogenated  
tallowamidoethyl-2-tallow imidazolinium  
methyl sulfate

The fabric softening mixtures of Formulations I, II and III above were heated and melted at approximately 80°C. Various silicones as indicated in Table V were added, with stirring, until the resulting mixture became hazy. At this point, the % silicone added was recorded as solubility of the silicone in the formulation. The results that were generated are summarised in Table V.

TABLE V

| Formulation<br>No. | PDMS <sup>1</sup> | Silicone Solubility (%)    |            |
|--------------------|-------------------|----------------------------|------------|
|                    |                   | Silicone E<br>(comparison) | Silicone F |
| I                  | 0.26              | 1.28                       | 4.70       |
| II                 | 0.34              | 0.69                       | 3.10       |
| III                | 0.39              | 1.69                       | 15.58      |

<sup>1</sup>PDMS = Polydimethylsiloxane, viscosity =  $1 \times 10^{-2} \text{ m}^2 \text{ s}^{-1}$  (10,000 cst)

Silicone F was significantly more soluble in Formulations I, II and III than PDMS.

EXAMPLE 8 (Comparison)

Various silicones both outside and within the scope of the invention as indicated in Table VI were incorporated into liquid fabric conditioning compositions. Fabric softening agents and silicones were mixed together at 80°C (above the melting point) and then dispersed into water at 60°C-80°C to form liquid compositions containing composite particles of the fabric softening component and the silicone.

The resulting compositions are summarised in Table VI.



TABLE VI

| Ingredients                | Sample A | B    | C    | D    | E    | F     | G     | H    |
|----------------------------|----------|------|------|------|------|-------|-------|------|
| Adogen 442                 | 7.3      | 7.3  | -    | -    | -    | 13.3  | -     | -    |
| Varisoft 475               | -        | -    | 10   | 10   | 10   | -     | -     | -    |
| Varisoft 445               | -        | -    | -    | -    | -    | -     | 11.7  | 11.7 |
| Neodol 23 <sup>1</sup>     | 0.94     | 0.94 | -    | -    | -    | -     | -     | -    |
| Siponic L7-90 <sup>2</sup> | 0.94     | 0.94 | -    | -    | -    | -     | -     | -    |
| Mineral Oil                | -        | -    | 10   | 10   | 10   | -     | -     | -    |
| Myristic acid              | -        | -    | -    | -    | -    | 1.25  | -     | -    |
| Stearic acid               | -        | -    | -    | -    | -    | -     | 3.5   | 3.5  |
| Silicone E<br>(comparison) | 0.119    | -    | 0.2  | -    | -    | 0.131 | 0.213 | -    |
| Silicone G<br>(comparison) | -        | 0.1  | -    | 0.2  | -    | -     | -     | -    |
| Silicone F                 | -        | -    | -    | -    | 0.2  | -     | -     | 1.9  |
| Water                      | 90.7     | 90.7 | 79.8 | 79.8 | 79.8 | 85.3  | 84.6  | 82.9 |

<sup>1</sup>Neodol 23 = Lauryl alcohol

<sup>2</sup>Siponic L-7-90 =  $C_{12}H_{25} - (OCH_2CH_2)_{12}OH$ , from Alcolac.

Samples C, D, E, G and H were further tested for their softening properties. Terry cloths were prewashed with a solution of Neodol 25-9 (alcohol ethoxylate from Shell Corp.) and  $Na_2CO_3$  to remove textile finishes on the surface, rinsed with the samples in a Tergometer and then line-dried. The cloth load was 20g per litre and the active concentration was 0.1g per litre of rinse liquid. The control was rinsed with only water. Using paired comparison, a panel of 20 judges assessed the softness of the treated cloth vs. control. All panelists preferred the treated cloths over the control in all tests.

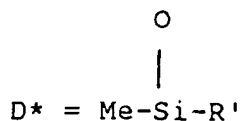
#### EXAMPLES 9-11 (Comparison)

The compatibility of various fabric softening agents with various silicones was determined by the SSCT. The entire concentration range up to 100% of the silicones was investigated. Samples that remained clear over the entire range of silicone concentration were labelled 'completely soluble'. For samples that became cloudy stability of the dispersions was ascertained and  $\alpha$  and  $\beta$  compatibility values were determined by the SSCT.

The silicones that were investigated are listed in Table II. In the silicone formulas of Table II.

M =  $Me_3SiO_{0.5}$

D =  $Me_2Si-O$



and R' is as indicated in Table II.

Table II

| Code | Formula   | R'                              | %CH <sub>2</sub> |
|------|---|---------------------------------|------------------|
| AA   | Polydimethylsiloxane<br>( $= 1 \times 10^{-3} \text{ m}^2 \text{ s}^{-1}$ (1000 cst)) | -                               | 0                |
| BB   | MD100D*5M   | C <sub>8</sub> H <sub>17</sub>  | 14               |
| CC*  | MD100D*5M   | C <sub>18</sub> H <sub>37</sub> | 28               |
| DD   | MD300D*20M  | C <sub>18</sub> H <sub>37</sub> | 28               |
| EE   | MD100D*10M  | C <sub>18</sub> H <sub>37</sub> | 43               |
| FF   | MD95D*24M   | C <sub>12</sub> H <sub>25</sub> | 57               |

\*Code CC, Formula MD100D\*5M is equivalent to Formula G of Example 8.

#### Example 9

In this example, mixtures of the silicones listed in Table II with mineral oil were investigated using the SSCT. The mineral oil used was Fished Light Mineral oil. The results that were generated are summarised in Table III.

Table III

| Compatibility with Mineral Oil |                        |                       |                     |
|--------------------------------|------------------------|-----------------------|---------------------|
| SILICONE                       | $\alpha$ COMPATIBILITY | $\beta$ COMPATIBILITY | COMPATIBLE (YES/NO) |
| AA                             | 1                      | 95                    | NO                  |
| BB                             | 4                      | 80                    | NO                  |
| CC                             | COMPLETELY SOLUBLE     |                       | YES                 |
| EE                             | COMPLETELY SOLUBLE     |                       | YES                 |
| FF                             | COMPLETELY SOLUBLE     |                       | YES                 |

As determined by the SSCT, silicones CC, EE and FF having the structural requirements and %CH<sub>2</sub> recited by the present invention form compatible mixtures with mineral oil.

#### Example 10

In this example, mixtures of the silicones listed in Table II with various cationic quaternary fabric softening agents were investigated using the SSCT.

The results that were generated are summarised in Tables IV, V and VI.

TABLE IVCompatibility with Varisoft 137<sup>1</sup>

| SILICONE | $\alpha$<br>COMPATIBILITY | $\beta$<br>COMPATIBILITY | COMPATIBLE<br>(YES/NO) |
|----------|---------------------------|--------------------------|------------------------|
| AA       | 2                         | 97                       | NO                     |
| BB       | 2                         | 98                       | NO                     |
| CC       | 2                         | 96                       | NO                     |
| EE       | 7                         | 93                       | YES                    |
| FF       | 7                         | 90                       | YES                    |

<sup>1</sup>Varisoft 137 = di(hydrogenated)tallow dimethyl ammonium methylsulfate from Sherex.

TABLE VCompatibility with Varisoft 445<sup>1</sup>

| SILICONE | $\alpha$<br>COMPATIBILITY | $\beta$<br>COMPATIBILITY | COMPATIBLE<br>(YES/NO) |
|----------|---------------------------|--------------------------|------------------------|
| AA       | 2                         | 97                       | NO                     |
| EE       | 10                        | 97                       | YES                    |
| FF       | -                         | 97                       | YES                    |

<sup>1</sup>Varisoft 445 = di(hydrogenated)tallow imidazolinium methylsulfate from Sherex.

TABLE VCompatibility with Varisoft 110<sup>1</sup>

| SILICONE | $\alpha$<br>COMPATIBILITY | $\beta$<br>COMPATIBILITY | COMPATIBLE<br>(YES/NO) |
|----------|---------------------------|--------------------------|------------------------|
|----------|---------------------------|--------------------------|------------------------|

|    |   |    |     |
|----|---|----|-----|
| AA | 1 | 98 | NO  |
| EE | 5 | 90 | YES |
| FF | 5 | 90 | YES |

<sup>1</sup>Varisoft 110 = methyl bis-(hydrogenated tallow amidoethyl)  
2-hydroxyethyl ammonium methysulfate from Sherex

Example 11

In this example, mixtures of the silicones listed in Table II with various nonionic fabric softening agents were investigated using the SSCT.

Results that were generated are summarised in Table VII, VIII, IX and X.

Table VIICompatibility with Neodol 45-7<sup>1</sup>

| SILICONE | $\alpha$<br>COMPATIBILITY | $\beta$<br>COMPATIBILITY | COMPATIBLE<br>(YES/NO) |
|----------|---------------------------|--------------------------|------------------------|
|----------|---------------------------|--------------------------|------------------------|

|    |   |    |     |
|----|---|----|-----|
| AA | 1 | 99 | NO  |
| BB | 1 | 99 | NO  |
| DD | 2 | 99 | NO  |
| FF | 5 | 93 | YES |

<sup>1</sup>Neodol 45-7 = ethoxylated fatty alcohol from Shell.

Table VIIICompatibility with Adogen 345D<sup>1</sup>

| SILICONE | $\alpha$<br>COMPATIBILITY | $\beta$<br>COMPATIBILITY | COMPATIBLE<br>(YES/NO) |
|----------|---------------------------|--------------------------|------------------------|
| A        | 2                         | 60                       | NO                     |
| B        | COMPLETELY SOLUBLE        |                          | YES                    |
| D        | COMPLETELY SOLUBLE        |                          | YES                    |
| E        | COMPLETELY SOLUBLE        |                          | YES                    |
| F        | COMPLETELY SOLUBLE        |                          | YES                    |

<sup>1</sup>Adogen 345D = di(hydrogenated)tallow dimethyl amine from Sherex.

Table IXCompatibility with PEG 600<sup>1</sup>

| SILICONE | $\alpha$<br>COMPATIBILITY | $\beta$<br>COMPATIBILITY | COMPATIBLE<br>(YES/NO) |
|----------|---------------------------|--------------------------|------------------------|
| AA       | 2                         | 99                       | NO                     |
| BB       | 2                         | 98                       | NO                     |
| DD       | 4                         | 95                       | NO                     |
| EE       | 4                         | 95                       | YES                    |
| FF       | 4                         | 95                       | YES                    |

<sup>1</sup>PEG 600 = Polyethylene Glycol

Table X

| Compatibility with isostearic acid |                        |                       |                     |
|------------------------------------|------------------------|-----------------------|---------------------|
| SILICONE                           | $\alpha$ COMPATIBILITY | $\beta$ COMPATIBILITY | COMPATIBLE (YES/NO) |
| AA                                 | 3                      | 95                    | NO                  |
| FF                                 | 3                      | 96                    | YES                 |

Examples 3-6 demonstrate that mutual compatibility between the fabric softening component and organosilicones may be easily determined by the SSCT and that the compatibility depends on the structure and %CH<sub>2</sub> content of the silicone as well as the particular fabric softening component employed in the mixture. Although silicone C was highly compatible (mutually soluble) with mineral oil in Example 3 and with Adogen 345D in Example 6, it was less compatible with Varisoft 137 of Example 4, i.e. a cloudy mixture was formed at 2% of silicone. However, silicone C was more compatible with Varisoft 137 in Example 4 than polydimethylsiloxane, since  $\beta$  compatibility was lower for silicone C than for polydimethylsiloxane. Results in Table VIII indicate that amines have the highest degree compatibility with organosilicones, since silicone B, which has the %CH<sub>2</sub> content of 14% and is not within the scope of this invention is still compatible with di(hydrogenated)tallow dimethyl amine. Silicones E and F, having a high %CH<sub>2</sub> content (43% and 57% respectively) were the most compatible with all softeners tested.

#### EXAMPLE 12 (Comparison)

Two fabric softening sheets, A and B were prepared as follows:

The ingredients of a fabric conditioning composition as listed below were mixed in the melt. 500g of the prepared fabric conditioning mixture was placed in the pan of a two-roll coating machine and coated onto a spun-bonded polyester non-woven material. The fabric softening articles thus manufactured contained about 1.6g of solidified softening composition. The articles of manufacture were then placed into a tumble dryer machine which already contained 2.2 kg of prewashed clothing, including terry towelling softness monitors. The fabrics were then tumble dried with the fabric softening article until dry and the softening benefit was evaluated by a 20 member panel.

Fabric Conditioning formulation for sheet A:

- 10% of a silicone not suitable for use in the present invention (silicone BB from Table II)
- 70% di(hydrogenated)tallow dimethyl ammonium methylsulfate
- 20% stearic acid.

Fabric Conditioning formulation for sheet B:

- 7% of silicone FF from Table II
- 70% di(hydrogenated tallow dimethyl ammonium methylsulfate
- 23% stearic acid.

#### Observations and results:

Sheet A - Due to the incompatible nature of the silicone, the silicone separated from the softening component during the coating process. The articles thus contained unknown amounts of the silicone.

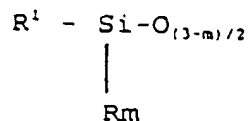
Sheet B - The compatible silicone FF and the softening component formed a compatible mixture which remained homogeneous during the coating process as it was transferred to the substrate indicating that the substrate was uniformly and evenly coated.

A 20 member panel judged the towelling monitors for both sheet A and sheet B to have superior softness vs. towels prepared in an identical fashion but dried without softener.

#### Claims

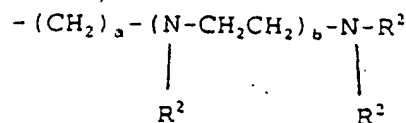
1. An article for conditioning fabrics, which provides for release of a fabric conditioning composition within an automatic laundry dryer at dryer operating temperatures, the article comprising a flexible substrate carrying thereon discrete composite particles consisting of a compatible mixture of:

- (a) at least 1% of a fabric softening component comprising a cationic quaternary ammonium salt; and  
 (b) an organosilicone having a %CH<sub>2</sub> content from 25% to 90% and having at least one unit of Formula A:

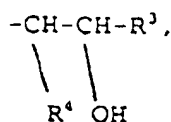


wherein m is a number from 0 to 2, R is a mono valent hydrocarbon radical and R<sup>1</sup> is

- (i) a unit of Formula A1

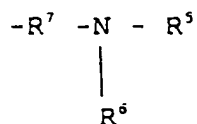


wherein a is a number of at least 1, b is a number from 0 to 10, R<sup>2</sup> is

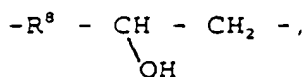


R<sup>3</sup> is a hydrocarbon radical having from 4 to 40 carbon atoms and R<sup>4</sup> is hydrogen or a hydrocarbon radical having from 1 to 40 carbon atoms; or

- (ii) a unit of Formula A2



wherein R<sup>5</sup> and R<sup>6</sup> are independently selected from hydrogen or a hydrogen radical having from 1 to 45 carbon atoms and at least one of R<sup>5</sup> and R<sup>6</sup> is a hydrogen radical having from 6 to 45 carbon atoms, R<sup>7</sup> is



wherein R<sup>8</sup> is a divalent organic radical having from 1 to 12 carbon atoms.

2. An article according to claim 1, characterised in that the %CH<sub>2</sub> content of the organosilicone(b) is 40% to 90%.
3. An article according to any preceding claim, characterised in that the composite particles consist of a mutually soluble mixture, which when in liquefied form give a clear homogeneous liquid.
4. An article according to any preceding claim, characterised in that R<sup>1</sup> includes from 8 to 18 carbon atoms.
5. An article according to any preceding claim, characterised in that a is 3 and b is 1.
6. An article according to any preceding claim, characterised in that R<sup>3</sup> includes from 8 to 18 carbon atoms.
7. An article according to any preceding claim, characterised in that R<sup>4</sup> is hydrogen.
8. An article according to any preceding claim, characterised in that m is one.
9. An article according to any preceding claim, characterised in that the amount of the organosilicone(b) is from 0.1% to 20% by weight of the particles.

10. An article as claimed in claim 9, characterised in that the amount of the organosilicone(b) is from 3% to 20% by weight of the particles.

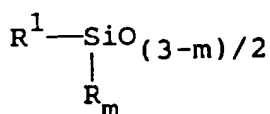
11. An article according to any preceding claim, characterised in that the flexible substrate is in a sheet configuration.

### Patentansprüche

1. Ein Artikel für die Gewebekonditionierung, welcher die Freisetzung einer gewebebedingenden Zusammensetzung innerhalb eines automatischen Wäschetrockners bei Trockner-Betriebstemperaturen vorsieht, wobei der Artikel ein flexibles Substrat einschließt, tragend darauf getrennte zusammengesetzte Teilchen, bestehend aus einer verträglichen Mischung von:

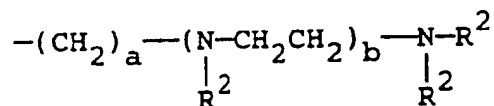
(a) zumindest 1 % einer gewebeerweichenden Komponente, einschließlich ein kationisches quaternäres Ammoniumsalz, und

(b) einem Organosilicon mit einem CH<sub>2</sub>-Prozentgehalt von 25 % bis 90 % und enthaltend zumindest eine Einheit der Formel A:

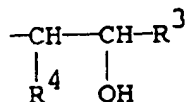


worin m eine Zahl mit einem Wert von 0 bis 2 ist, R einen einwertigen Kohlenwasserstoffrest bedeutet und R<sup>1</sup>

(i) eine Einheit der Formel A1

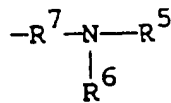


ist, worin a eine Zahl mit einem Wert von zumindest 1 bedeutet, b eine Zahl von 0 bis 10 ist, R<sup>2</sup>

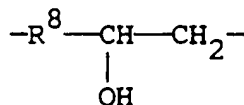


bedeutet, worin R<sup>3</sup> ein Kohlenwasserstoffrest mit 4 bis 40 Kohlenstoffatomen und R<sup>4</sup> Wasserstoff oder ein Kohlenwasserstoffrest mit 1 bis 40 Kohlenstoffatomen ist, oder

(ii) eine Einheit der Formel A2



bedeutet, worin R<sup>5</sup> und R<sup>6</sup>, unabhängig, aus Wasserstoff oder einem Kohlenwasserstoffrest mit 1 bis 45 Kohlenstoffatomen ausgewählt sind und zumindest einer der Reste R<sup>5</sup> und R<sup>6</sup> ein Kohlenwasserstoffrest mit 6 bis 45 Kohlenstoffatomen ist, worin R<sup>7</sup>



bedeutet, worin R<sup>8</sup> ein zweiwertiger organischer Rest mit 1 bis 12 Kohlenstoffatomen ist.

2. Ein Artikel nach Anspruch 1, **dadurch gekennzeichnet**, daß der CH<sub>2</sub>-Prozentgehalt des Organosilicons (b) 40 % bis 90 % beträgt.

3. Ein Artikel nach einem der vorstehenden Ansprüche, **dadurch gekennzeichnet**, daß die zusammengesetzten



Teilchen aus einer wechselseitig löslichen Mischung bestehen, welche, sobald in verflüssigter Form, eine klare homogene Flüssigkeit liefert.

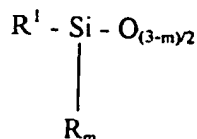
- 5 4. Ein Artikel nach einem der vorstehenden Ansprüche, **dadurch gekennzeichnet**, daß R<sup>1</sup> von 8 bis 18 Kohlenstoffatome enthält.
5. Ein Artikel nach einem der vorstehenden Ansprüche, **dadurch gekennzeichnet**, daß a einen Wert von 3 und b einen Wert von 1 aufweist.
- 10 6. Ein Artikel nach einem der vorstehenden Ansprüche, **dadurch gekennzeichnet**, daß R<sup>3</sup> von 8 bis 18 Kohlenstoffatome enthält.
7. Ein Artikel nach einem der vorstehenden Ansprüche, **dadurch gekennzeichnet**, daß R<sup>4</sup> Wasserstoff ist.
- 15 8. Ein Artikel nach einem der vorstehenden Ansprüche, **dadurch gekennzeichnet**, daß m den Wert 1 aufweist.
9. Ein Artikel nach einem der vorstehenden Ansprüche, **dadurch gekennzeichnet**, daß die Menge des Organosilicons (b) im Bereich von 0,1 bis 20 Gewichtsprozent der Teilchen liegt.
- 20 10. Ein Artikel nach Anspruch 9, **dadurch gekennzeichnet**, daß die Menge des Organosilicons (b) im Bereich von 3 bis 20 Gewichtsprozent der Teilchen liegt.
11. Ein Artikel nach einem der vorstehenden Ansprüche, **dadurch gekennzeichnet**, daß das flexible Substrat in einer Blattkonfiguration vorliegt.

#### Revendications

- 30 1. Un article pour le conditionnement de tissu qui fournit la libération d'une composition de conditionnement de tissu à l'intérieur d'un sèche-linge automatique à des températures de séchage, l'article comprenant un substrat flexible sur lequel se trouvent des particules composites discrètes composées d'un mélange compatible de :

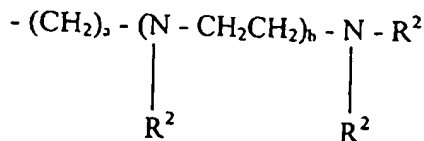
(a) au moins 1 % d'un composant adoucissant le tissu comprenant un sel d'ammonium quaternaire cationique ;  
et

35 (b) une organosilicone comprenant une teneur en CH<sub>2</sub> (%) comprise entre 25 % et 90 % et présentant au moins une unité de formule A :

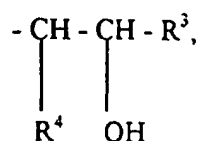


45 où m est un nombre compris entre 0 et 2, R est un radical hydrocarbure monovalent et R<sup>1</sup> est

(i) une unité de Formule A1

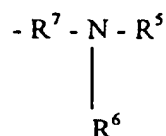


55 où a est un nombre au moins égal à 1, b est un nombre compris entre 0 et 10, R<sup>2</sup> est

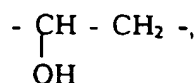


R<sup>3</sup> est un radical hydrocarbure présentant 4 à 40 atomes de carbone et R<sup>4</sup> est de l'hydrogène ou un radical hydrocarbure présentant 1 à 40 atomes de carbone ; ou

(ii) une unité de Formule A2



où R<sup>5</sup> et R<sup>6</sup> sont indépendamment sélectionnés parmi de l'hydrogène ou un radical hydrogène présentant 1 à 45 atomes de carbone, et au moins un des groupes R<sup>5</sup> et R<sup>6</sup> est un radical hydrogène présentant 6 à 45 atomes de carbone, R<sup>7</sup> est - R<sup>8</sup>



où R<sup>8</sup> est un radical organique divalent présentant 1 à 12 atomes de carbone.

2. Un article selon la Revendication 1, caractérisé en ce que la teneur en CH<sub>2</sub> (%) de l'organosilicone (b) est comprise entre 40 % et 90 %.
3. Un article selon l'une des Revendications précédentes, caractérisé en ce que les particules composites se composent d'un mélange réciproquement soluble qui, sous forme liquéfiée, forme un liquide homogène clair.
4. Un article selon l'une des Revendications précédentes, caractérisé en ce que R<sup>1</sup> comprend 8 à 18 atomes de carbone.
5. Un article selon l'une des Revendications précédentes, caractérisé en ce que a est égal à 3 et b est égal à 1.
6. Un article selon l'une des Revendications précédentes, caractérisé en ce que R<sup>3</sup> comprend 8 à 18 atomes de carbone.
7. Un article selon l'une des Revendications précédentes, caractérisé en ce que R<sup>4</sup> est de l'hydrogène.
8. Un article selon l'une des Revendications précédentes, caractérisé en ce que m est égal à un.
9. Un article selon l'une des Revendications précédentes, caractérisé en ce que la quantité d'organosilicone (b) est comprise entre 0,1 % et 20 % en poids des particules.
10. Un article tel que revendiqué dans la Revendication 9, caractérisé en ce que la quantité d'organosilicone (b) est comprise entre 3 % et 20 % en poids des particules.
11. Un article selon l'une des Revendications précédentes, caractérisé en ce que le substrat flexible se présente sous forme d'une feuille.